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THE SO-CALLED HALLOYSITE OF JONES FALLS, MARYLAND[1]

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Widely distributed specimens from the gneiss quarries along Jones Falls in the city of Baltimore, Maryland, contain zeolites coating narrow fractures. These include stilbite, chabazite of the variety called haydenite, and heulandite of the crystal habit first described under the name beaumontite. The beaumontite-heulandite is younger than the others and rests upon them. It is frequently noted that the older generation of stilbite and haydenite are altered and to a greater or less extent replaced by a fine steel or lead gray to olive green mineral of clayey consistency which may form pseudomorphs after the rhombohedral crystals of haydenite. This soft clayey mineral may also form thin coatings on otherwise bright and uncorroded haydenite crystals.

In the specimen investigated the clayey mineral was of an olivine-green color and occurred as masses of irregular form between and on rhombohedral crystals of haydenite up to 5 mm. on an edge. Although rusty and partly coated with small beaumontite crystals, where fractured the haydenite is light brownish-pink and is comparatively unaltered, although in other specimens rhombohedral pseudomorphs after haydenite now consist entirely of the secondary mineral. In this specimen the greenish areas possess a radiating structure clearly inherited from the stilbite. The mineral rusts brown on the outside and is filled with contraction cracks. When pieces are placed in water they disintegrate somewhat by the splitting off of curved fragments after the manner of an "aquacreptite" and become more translucent and more yellowish in color. The mineral does not become plastic when moist.

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[1] Published by permission of Dr. Charles D. Walcott, Secretary of the Smithsonian Institution. This paper is the fifth preliminary paper on subjects of interest investigated in connection with the preparation of a bulletin on the minerals of Maryland in cooperation with the Maryland State Geological survey.

This mineral is specifically mentioned by Williams[2] who states that it is often called chlorite but is not chlorite but halloysite, rendered impure by admixture of silica. He describes it as a compact, apparently amorphous mineral of a light blue color, sometimes occurring in connection with the haydenite, which turns brown on exposure to the air. An analysis, made by Bayley, is given which probably represents a very impure sample.

In the present investigation a fairly pure sample was prepared by digging the mineral out with a needle point, screening it to uniform size after grinding, and separating with an electromagnet and heavy solutions. This was analyzed with the results and ratios given in the following table:

ANALYSIS AND RATIOS OF "HALLOYSITE"

CONSTITUENTS	PER CENT	RATIOS		
SiO_2	48.92	.815	.815	1.00×3
Al_2O_3	18.96	.186	.241	.89×1
Fe_2O_3	8.75	.055		
FeO	1.09	.015	.101	.92×2/5
CaO	1.68	.030		
MgO	2.24	.056		
H_2O	19.00	1.055	1.055	.97×4
Total	100.64			

The ratios of the above analysis lead to the formula: $2/5\ R''O.R'''_2O_3.3SiO_2.4H_2O$. The bivalent bases representing less than half a molecule may be dropped or may be included with the water. In the former case the formula may be simplified to $Al_2O_3.3SiO_2.4H_2O$. This formula is exactly that obtained by a recalculation of the analysis of the "leverrierite" of Wherry and Larsen[3] from Beidell, Colo., and from the writer's analysis of "leverrierite" from Idaho.[4] It is evident that this mineral from Maryland is essentially identical, chemically, with these gouge clays.

Under the microscope this material shows a very fine aggregate crystalline-micaceous structure and greenish-brown color. It is obvious that it was deposited in a colloidal form but it is now completely crystalline. It is not fibrous with the structure which is commonly called metacolloidal, for example, like chalcedony. No pleochroism could be detected in the fine aggregates nor could any interference figure be obtained from such fine-grained material.

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[2] George N. Williams: Notes on the minerals occurring in the neighborhood of Baltimore. *Johns Hopkins Univ. Publications*, Baltimore, **1887**.

[3] Esper S. Larsen and Edgar T. Wherry: Leverrierite from Colorado. *J. Wash. Acad. Sci.*, **7**, 208-217 (1917).

[4] Earl V. Shannon: Notes on the mineralogy of three gouge clays from precious metal veins. *Proc. U.S. Nat. Mus.*, **62**, Art. 15, (1922).

There can remain little doubt that this mineral is not halloysite, the isotropic equivalent of kaolinite but, it is one of the high-water clay group identical with the gouge clays and, as indicated by unpublished work, with the Arkansas bentonite. It now appears that the name leverrierite was inappropriate for this compound but it has not, as yet, been decided just what mineral name should be applied to it. Its occurrence in the present specimens as an alteration product of zeolites is, so far, unique and quite different from its other modes of origin.

ANTLERITE FROM CHUQUICAMATA, CHILE

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In a lot of unlabelled specimens of copper ore from Chuquicamata, Chile, recently sent by Mr. Juan L. Carrasco, there was found in abundance a dark green mineral occurring in thin veins in a sericitized granitic rock. In one specimen it occurs with gypsum and, for the most part, not associated with other copper minerals. It was supposed that the green material was brochantite, atacamite, or a mixture of the two, since these have been reported[1] as among the principal ore minerals of this locality. However, chemical analyses and determination of optical properties show conclusively that it is antlerite ($3CuO.SO_3.2H_2O$) and not brochantite ($4CuO.SO_3.3H_2O$).

Crystals of antlerite from the same locality have recently been described and drawn by Ungemach,[2] who found from measurements on very good crystals that they are orthorhombic with an axial ratio of $a:b:c=0.5038:1:0.6868$. The material examined by the writers was in part well crystallized but measurements were carried out only far enough to locate the crystallographic c axis for determination of the optical orientation. The habit of the crystals varies; some are flattened parallel to (010), which is the direction of the highly perfect cleavage; others are about equally developed along the three axes. The greatest thickness of any of the veins in the specimens available is only about 2 or 3 mm.; some of the flat crystals have a thickness about equal to this and a much greater length and width.

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[1] Palache and Warren: *Am. J. Sci.*, **26,** 345 (1908).

J. A. Hendricks: *Univ. of Cal. Publications,* **14,** 75-84 (1922).

[2] *Bull. Soc. Min. France,* **47,** 124 (1924).

The optical properties of the antlerite were determined by J. H. C. Martens as follows: refractive indices, $\alpha = 1.730 \pm .010$, $\lambda = 1.786 \pm .010$; optical character positive, $2V$ moderate; orientation $X = b$, $Y = c$, $Z = a$; the obtuse bisectrix therefore is perpendicular to the (010) cleavage; pleochroism rather strong, X = yellowish green, $Y = Z$ = bluish green. Some parts of the antlerite appear in mass darker and more opaque than others, but microscopic examination of thin fragments shows that there is no variation in the mineral itself but that the lesser degree of transparency is caused by small inclusions of colorless or nearly colorless isotropic material of low refractive index, probably air or water. The hardness of antlerite is about 3, a little less than that of brochantite.

Duplicate analyses made by L. F. Audrieth on carefully selected material containing no impurities except some fragments of the wall rock, which appear in the analysis as insoluble material, and less than one per cent by volume of the small isotropic inclusions mentioned above, gave the following results.

ANALYSES OF ANTLERITE, $3CuO.SO_3.2H_2O$

	I	II	Mean of I and II	Theoretical
CuO	66.48	66.21	66.34	67.28
SO_3	22.38	22.27	22.32	22.56
H_2O	10.75	10.29	10.52	10.16
Volatile (H_2O+SO_3)	33.07	.....	.....	.....
Insol. in HCl	0.87	0.89	0.88	.....
Cl	none	.....	.....	.....
			100.06	100.00

Water in analysis I was determined by subtracting the average SO_3 content from the total volatile matter. The volatile matter was determined by heating a sample to constant weight in a platinum crucible over a blast lamp. Water in analysis II was determined as loss on ignition, using CaO to retain the SO_3 and taking care to avoid absorption of CO_2 from the air by the CaO.

The loss of weight at 120°C. is negligible, although the mineral decrepitates noticeably at higher temperature indicating that water may be occluded within the crystals.

Spectrographs of the antlerite indicate the presence of small amounts of silicon, aluminum and titanium, probably in the form of oxides and included under the insoluble matter in the above analyses. Traces of calcium, iron and silver are also present in quantities too small to be detected by other analytical methods (less than 0.01 per cent).

In the same collection of specimens from Chuquicamata, chalcanthite and natrochalcite were also found, the latter in distinct crystals of the type recorded by Palache and Warren, and in such abundance that it is probably one of the important ore minerals.

The recent recognition of antlerite as occurring in this deposit, evidently in large amounts, suggests that much of what has been called brochantite may really be antlerite, since the two minerals are similar in general appearance and their qualitative chemical reactions are the same.

THE FORM OF REPLACEMENT CRYSTALS

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Epidote crystals introduced during the replacement of a rock mass[1] have been found elongated parallel to the *c* axis, a direction along which elongation of epidote is very rare. The sections contain a great many of these epidote crystals which are mostly microscopic in size, but in every case the elongation was found to be parallel to the *c* axis rather than along *b*, the common direction of elongation. The development of such a rare crystal form under replacement conditions warrants an explanation since it may make clear certain features of replacement processes.

The mechanism of replacement[2] and the probable nature of the introduced material will not be discussed here beyond a few brief comments pertinent to the subject of this paper.

The positions occupied by material favorable to the reaction with the introduced substance no doubt determined the positions in which the epidote crystals developed. The material introduced may have entered chiefly by diffusion through amorphous grain boundaries, or along cleavage planes, or directly through the crystal lattice itself. The velocity of such diffusion should be greatest through amorphous grain boundaries, less along cleavages and least through a crystal lattice. Replacement, especially at its inception, is commonly observed along grain boundaries working into the replaced mineral along cleavages.

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[1] Observed in thin sections of rocks from Courtland, Arizona, in the possession of Professor L. C. Graton, Harvard University.

[2] The Mechanism of Replacement and Recrystallization: Ernest E. Fairbanks, *Econ. Geol.*, forthcoming article.

Replacements appear to be volume for volume in nature, as pointed out by Lindgren. Changes taking place *within* the crystal lattice should serve to explain such relationships. Upon first thought it might appear as though the introduction of material into a rock mass would tend to produce an increase in volume. Such introduction is, undoubtedly, attended by loss of material as a result of a diffuse cycle which is initiated by the entrance of the foreign material into the rock mass.

In the case of a portion of the rock material reacting with the introduced substance it would appear as though volume relations should vary. It is known, however, that the molecular volume of a compound is not, as a rule, the sum of the atomic volumes of its component elements, a notable contraction usually occurring on the combination of the elements. This contraction is due to the difference in the crystal structures of the solid elements and the solid compound. Therefore, it would appear as though an actual decrease in volume should occur as a result of replacement in some cases. A compensating influence is afforded by attendant increase in volume similar in magnitude to that of the decrease as a result of the production of solid solutions. The substitution of atoms, as shown from X-ray study, involves a distortion of the crystal lattice. The crystal lattice of a solid solution continues to possess the lattice of the solvent up to a certain concentration of solute producing an expansion of the lattice, which, in the case of 8 per cent Mg replacing Al, amounts to an increase from 4.05 to 4.10 Å.

If the foregoing facts are significant in the control of volume relationships, a relation should be expected between the two rare cases (1) when an increase in volume takes place on combination, and (2) when a decrease in volume is produced by atom substitution during solid solution formation.

Volume relations are not necessarily preserved under near surface replacements indicating that pressure is an important factor in the preservation of volume relationships.

Passing over the subject of volume relationships with this brief hint regarding the nature of the process, we have yet to consider the factors which tend to produce unusual crystal elongation. Crystallization velocities vary at various crystal faces.[3] Normally the velocity along the *b* axis of epidote is the greatest. In the presence of foreign material adsorption[4] of such material may also be greatest upon the faces developed at the extremities of the *b* axis. Now adsorption of foreign material is known to impede the crystallization velocity (C.V.). If this inhibition reduces the velocity along *b* until it is less than along the *c* axis, the result will be that the crystal will grow more rapidly along *c* and thus become elongated as found in the study of the thin sections.

The mechanism of the inhibiting effect of adsorbed material upon the C.V. may be given briefly as follows: molecules or atoms form an adsorption layer upon a surface of a crystal lattice. The molecules or atoms of such layers are not permanently attached but are considered to constantly shift away from the lattice and back. The retention of molecules requires time and during this period some other surface may be building more rapidly. Hence a change in original velocities may be produced.

The attractive force of an atom is a factor which likewise may be important in producing changes in C. V. Adsorption layers may be as thick as 20 or 50 molecules in some instances, while in other cases it may have the thickness of a single molecule. The distance to which this attraction can extend is a debatable subject.

Considering epidote through its entire period of formation we would expect foreign material to be adsorbed upon some surface of the unit-cell the moment such a cell is formed, providing conditions which favor adsorption are satisfied. Here we are dealing with anisotropic media and adsorption can affect the crystallization velocities of the crystal axes while such action is impossible if the media remains isotropic.[5]

[3] The Effect of Various Factors Upon the Velocity of Crystallization of Substances from Solution: John D. Jenkins, *J. Am. Chem. Soc.*, **47**, No. 4, April 1925, pp. 903-922. Jenkins determined the velocities of crystallization by means of two methods, (1) a rapid refractometric method and (2) a thermometric method.

[4] Adsorption is here considered to be a manifestation of both the causal factors of chemical combination and true solution i.e., molecular attraction and the kinetic motion of the molecules, chemical combination being entirely due to the attractive forces between the atoms or molecules, while true solution is a manifestation of the tendency of the thermal agitation of the molecules to produce a chaotic mixture.

[5] Further discussion of crystal modification with special attention to the mechanism as given by Valeton is being prepared for a future article.

Only a small quantity of foreign material need be adsorbed to control the crystallization. Analyses of various epidotes show a varying amount of impurities. The greater portion of this foreign material was occluded during the process of crystallization and was not added by the process of adsorption.

The elongation of epidote crystals along the *b* axis is believed to be satisfactorily explained as a result of the reaction of introduced material with rock material amenable to such reactivity. During the process of crystallization, due to adsorption of foreign material, the growth of the crystals along the *b* axis is retarded to a greater extent than along the *c* axis.

Lindgren's[6] contention, that replacement is essentially a process of adsorption and super-saturation, receives, then, additional support from the foregoing observations, with regards to the importance of adsorption.

[6] Mineral Deposits, p. 26, New York, 1919.

A NEW THEORY OF THE COMPOSITION OF THE ZEOLITES. PART IV

(*Continued from page 152*)

A. N. Winchell, *University of Wisconsin*

5. The Composition of Heulandite and Related Zeolites

Doelter[1] has collected 47 analyses of heulandite and three more have been published since the date of his compilation. However, many of these are too old to be of high quality, and some were made on impure material. In heulandite, as in other zeolites, the ratio between Al_2O_3 and $\ddot{R}O+R_2O$ is very nearly constant and very nearly unity. Excluding therefore a few analyses because this ratio is unsatisfactory, the "superior" analyses of heulandite are the following:

Doelter's Nos. 11, 19, 23, 25, 37, 38, 40, 41, and 47.
A. L. Parsons:[2] Heulandite from Digby Cut, Nova Scotia.
K. H. Scheumann:[3] Heulandite from Berufiord, Iceland.

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[1] Hdb. Mineralchemie, **II**, 3, (1921), p. 171.

[2] *U. Toronto Studies, Geol. Ser.*, **14**, (1922), p. 52.

[3] *Ber. Sachs. Akad. Wiss.*, Leipzig, **LXXIII**, (1921), p. 1; *Zeit. Kryst.*, **LVII**, (1923), p. 657.

In spite of the fact that the differences in composition are not very great it may be useful to study these analyses by the method recently described.[4] However, in this case the corners of the square must be more highly siliceous than in the case of thomsonite, or even natrolite, since the points for heulandite cannot be plotted on the square as used for those minerals. As a matter of convenience the following molecules are chosen as the corners of the square:

	Lime	Soda
More siliceous	$CaAl_2Si_{14}O_{32}.Aq.$	$Na_2Al_2Si_{14}O_{32}.Aq.$
Less siliceous	$Ca_3Al_6Si_{10}O_{32}.Aq.$	$Na_6Al_6Si_{10}O_{32}.Aq.$

It is evident that the ratio between Al_2O_3 and $CaO+Na_2O$ is unity at all points on the square, and it is easy to see that the horizontal axis expresses the $CaO:Na_2O$ relation while the vertical axis expresses the $Al_2O_3:SiO_2$ relation. It will be readily understood that CaO and Na_2O in the figure stand for $CaO+MgO+SrO+BaO$ and Na_2O+K_2O, respectively. It is easily seen that the lower half of this square is the same as the upper half of the square used for natrolite.

The results of calculating the "superior" analyses into these molecules and plotting them on the square are shown in Fig. 5, on which are also shown points representing similarly selected analyses of epistilbite, mordenite and brewsterite, which are isomorphous with heulandite. The variations in composition in heulandite are not sufficiently great to put the proper interpretation beyond question, but they are as favorable to the theory that the composition of heulandite varies like that of feldspars as they are to any other theory, that is, the points should lie along a curve if the composition varies as in the feldspars, while, if it varies, as is commonly supposed in all zeolites, so that Na_2 takes the place of Ca the points should lie along a horizontal line. The writer has already pointed out that such a variation in composition as would be expressed in the diagram by a horizontal line can be produced in zeolites (already crystallized) by artificial means, and it seems probable that some variations of this kind may occur in nature *after formation* of the zeolite crystals.

It is not clear precisely what ratio of Ca+Na atoms to oxygen atoms is found in heulandite, but the writer would suggest that the mineral may be represented approximately as isomorphous mixtures in variable proportions of the molecules: $NaCa_5Al_{11}Si_{29}O_{80}.25H_2O$ and $Na_2Ca_4Al_{10}Si_{30}O_{80}.25H_2O$.

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[4] A. N. Winchell: The Composition of Thomsonite, *Am. Min.*, **X,** (1925), p. 90.

In these formulas the writer is merely expressing the ordinary view regarding the water. Neither the amount nor the condition of the water is considered in this discussion, except that it is assumed that the water takes no part in the main formula. This assumption is in harmony with the fact that the water can be driven out without destroying the zeolite crystal.

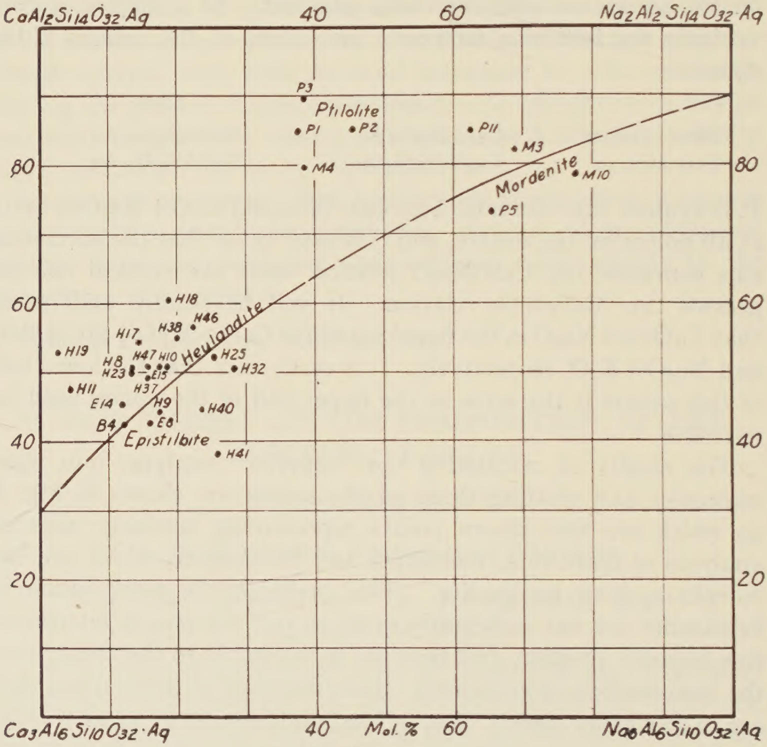

Figure 5
The Composition of Heulandite and Related Zeolites

Tschermak[5] has recently made it clear that mordenite and epistilbite (as well as brewsterite) belong to the heulandite group by adopting a new orientation for epistilbite, so that the ratios become:

Mordenite	$a:b:c=0.401:1:0.428$	$\beta=88°31'$
Heulandite	$a:b:c=0.404:1:0.429$	$\beta=88°34'$

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[5] *Sitz. Akad. Wiss. Wien,* **CXXVI,** (1917), p. 601.

Epistilbite $a:b:c=0.419:1:0.432$ $\beta=89°20'$
Brewsterite $a:b:c=0.405:1:0.420$ $\beta=86°20'$

Mordenite is so rare that only three satisfactory analyses have been found. These are shown on the figure (M10 the analysis by E. W. Todd published by T. L. Walker: *Toronto U. Studies, Geol. Ser.*, **14**, (1922), p. 59; the others are Doelter's Nos.). Except for analysis M4, mordenite belongs to the same series as heulandite, and is nearly free from lime, the formula being roughly $Na_4Ca_2Al_8Si_{32}O_{80}.25H_2O$.

Epistilbite is chemically nearly the same as one end member of the heulandite series: no modern analyses are available, but two, carefully selected from the old analyses, are shown on Fig. 5. Therefore the composition of epistilbite may be indicated as $NaCa_5Al_{11}Si_{29}O_{80}.25H_2O$.

Brewsterite is rare and only one superior analysis is on record. So far as shown by the diagram it is practically the same as epistilbite, but it is characterized by the presence of much strontium and some barium.

So far as the optic properties of the epistilbite-heulandite-mordenite series are concerned, the mean index of refraction and the birefringence seem to decrease regularly with increase of soda and silica, but the sign and optic orientation do not seem to be those that should belong to such a series. However, when it is remembered that small temperature changes produce marked changes in these properties, as shown by Rinne for stilbite[6] and heulandite,[7] and that heulandite and epistilbite both become orthorhombic with similar optic orientations at moderate temperatures, the theory that they belong to a single isomorphous series does not seem unreasonable.

It may be noted that, if brewsterite is isomorphous with heulandite and epistilbite, as seems clearly to be the case from the chemical and crystallographic data, the index of refraction in the literature for the former[8] ($n=1.45$) must be incorrect since its composition as compared with that of the latter would require a higher index than possessed by heulandite and epistilbite (namely 1.50 and 1.51). It may be suggested that the index of brewsterite should be 1.55, the 1.45 being perhaps a typographical error.

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[6] *N. Jahrb. Min.*, (1897), **I,** p. 41.

[7] *Sitz. preus. Akad. Wiss. Berlin,* **XLVI,** (1890), p. 1183.

[8] Minéraux des Roches, **1888,** p. 301.

Ptilolite is chemically closely related to mordenite, but Böggild[9] has shown that it is normally orthorhombic. The analysis (P11 in Fig. 5) by Prof. Kossmat recently published by Tschermak[10] suggests that the mineral has the composition $Na_3Ca_2Al_7Si_{33}O_{80}.25H_2O$, differing from mordenite in having 5(Na+Ca) atoms, instead of 6, for 80 atoms of oxygen. However, Lindström's analysis (P5 in Fig. 5) of ptilolite from Teigarhorn, Iceland,[11] has the same ratio as found in mordenite; it may be significant that Böggild found the mineral in this occurrence to differ optically from all others, being apparently monoclinic like mordenite. There are three other good analyses of ptilolite, two by Eakins[12] (P1 and P2 in Fig. 5) and one by Colomba[13] (P3 in Fig. 5). This is the first instance discovered among the zeolites in which the data indicate a series by substitution of two atoms of Na for one of Ca. The analyses are not numerous and the condition is perhaps due to change in composition after formation of the crystals, as previously suggested.

6. Comparative Study and Summary

A summary of the composition of the zeolites as determined in this study may be arranged as follows:

Thomsonite	$Na_4Ca_8Al_{20}Si_{20}O_{80}.25H_2O$ to $Na_7Ca_5Al_{17}Si_{23}O_{80}.20H_2O$.
Gmelinite	$Na_4Ca_2Al_7Si_{13}O_{40}.20H_2O$ to $Na_6Al_6Si_{14}O_{40}.20H_2O$
Levynite	$Na_4Ca_4Al_9Si_{16}O_{50}.25H_2O$
Chabazite	$Ca_7Al_{14}Si_{26}O_{80}.40H_2O$ to $(Na, K)_4Ca_3Al_{10}Si_{30}O_{80}.40H_2O$
Phillipsite	$K_4Ca_7Al_{18}Si_{22}O_{80}.40H_2O$ to $K_6Ca_5Al_{16}Si_{24}O_{80}.30H_2O$
Stilbite	$Na_3Ca_9Al_{21}Si_{59}O_{160}.60H_2O$
Harmotome	$KBa_5Al_{11}Si_{29}O_{80}.25H_2O$
Natrolite	$Na_2Al_2Si_3O_{10}.2H_2O$
Mesolite	$Na_2Ca_2Al_6Si_9O_{30}.8H_2O$
Scolecite	$CaAl_2Si_3O_{10}.3H_2O$
Edingtonite	$BaAl_2Si_3O_{10}.3H_2O$
Gismondite	$KCa_{10}Al_{21}Si_{19}O_{80}.40H_2O$ to $K_4Ca_7Al_{18}Si_{22}O_{80}.36H_2O$
Laumontite	$Ca_7Al_{14}Si_{26}O_{80}.25H_2O$ to $Na_2Ca_5Al_{12}Si_{28}O_{80}.25H_2O$
Heulandite	$NaCa_5Al_{11}Si_{29}O_{80}.25H_2O$ to $Na_2Ca_4Al_{10}Si_{30}O_{80}.25H_2O$
Mordenite	$Na_4Ca_2Al_8Si_{32}O_{80}.25H_2O$?
Epistilbite	$NaCa_5Al_{11}Si_{29}O_{80}.25H_2O$
Brewsterite	$Na(Sr,Ba,Ca)_5Al_{11}Si_{29}O_{80}.25H_2O$
Ptilolite	$Na_3Ca_2Al_7Si_{33}O_{80}.25H_2O$?

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[9] *Kgl. Danske Vid. Selsk. Math. fys. Meddel.*, **IV**, 8, (1922).

[10] *Sitz. Akad. Wiss. Wien*, **CXXVI**, (1917), p. 547.

[11] *Geol. För. Förh. Stockholm*, **XX**, (1907), p. 106.

[12] *Am. J. Sci.*, **XXXII**, (1886), p. 117, and **XLIV**, (1898), p. 96.

[13] *Zeit. Kryst.*, **XL**, (1904), p. 100.

The most striking feature of these formulas is their remarkable complexity. It is possible that in some cases more accurate information will permit some simplifications, but, in general, this will probably not be considerable, and it must be concluded that the zeolites are remarkable in one other respect, besides those mentioned at the beginning of this discussion, namely, in the extraordinary complexity of their formulas.

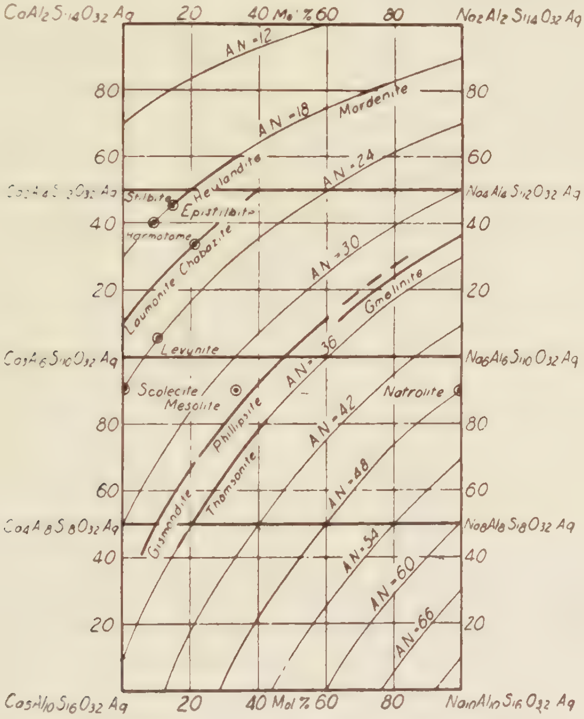

Figure 6

Summarized Composition of the Zeolites

AN (alkali number) = number of (Ca+Na) atoms to 240 oxygen atoms.

However, the preceding discussion shows that the zeolites are like the feldspars in their composition in certain important respects: namely,

1. The Al_2O_3:$CaO+Na_2O$ ratio is always unity in the whole zeolite group.
2. The Al+Si:O ratio is always 1:2 in the whole group.
3. In any given strictly isomorphous part of the zeolite group the Ca+Na:O ratio is constant.

4. The composition of all the zeolites can be expressed very simply in a suitable graph, in which the composition of certain zeolites is shown by points, while that of others varies along simple lines, as shown in Fig. 6.

If the number of (Ca+Na) atoms in each portion of the zeolite space lattice containing 240 oxygen atoms be called the alkali number then it appears from the preceding study that the following tabulation is possible:

Name	Alkali number
Ptilolite	15?
Mordenite	18
Heulandite	18
Epistilbite	18
Brewsterite	18
Stilbite	18
Harmotome	18 or 21?
Laumontite	21
Chabazite	21
Levynite	24
Scolecite	24
Edingtonite	24
Feldspar	30
Mesolite	32
Gismondite	33
Phillipsite	33
Gmelinite	34 or 36?
Thomsonite	36
Natrolite	48

This suggests a new study of the crystallographic relationships of the zeolites, but the subject will not be pursued farther here.

The writer believes that the preceding study of the zeolites has shown that:

1. Thomsonite is certainly an isomorphous series of the feldspar type.

2. Natrolite, mesolite and scolecite are three independent zeolites of fixed composition, which are not isomorphous in the sense of forming intermediate mix-crystals. They have similar crystal forms, but unlike space lattices, as shown both by their inability to form mutual crystal solutions and by their differences of symmetry.

3. Laumontite is probably a short isomorphous series of the feldspar type; it is chemically related to chabazite, but contains less water.

4. Gismondite is probably a short isomorphous series of the feldspar type; it is chemically closely related to phillipsite.

5. Chabazite probably forms a short isomorphous series of the feldspar type; it is chemically related to laumontite but contains more water.

6. Gmelinite probably forms an isomorphous series of the feldspar type; the variation of some crystals toward the composition of chabazite is believed to be due to change in composition after formation of the crystals just as "permutite" and many natural zeolites are known to change composition in various solutions under valence-control.

7. Levynite is unlike chabazite in composition and may be of fixed formula.

8. Phillipsite is an isomorphous series of the feldspar type, though variations in composition under valence-control, believed to be secondary, as in the case of gmelinite, are also known. Phillipsite is chemically closely related to gismondite.

9. With three exceptions, perhaps misnamed, stilbite is an independent zeolite of fixed composition; it is related to epistilbite and heulandite in composition.

10. Harmotome is perhaps of fixed composition; it is related to stilbite, but is rich in barium.

11. Heulandite varies in composition rather lawlessly, but may be referred reasonably to an isomorphous series (with mordenite) of the feldspar type.

12. Epistilbite belongs at one end of the heulandite series.

13. Brewsterite is like epistilbite in composition except that strontium and barium replace much of the calcium of the latter.

14. Disregarding one analysis, as perhaps due to secondary changes under valence-control, mordenite belongs, with heulandite, to a discontinuous (?) isomorphous series of the feldspar type.

15. Ptilolite seems to form an isomorphous series in which 2Na takes the place of Ca, or *vice versa*, but analyses are few and the variations may be due to changes after formation of the crystals, such as can be produced artificially in several zeolites.

16. The general conclusion which seems justified may be stated as follows:

With a few exceptions which can reasonably be explained as due to secondary changes after formation of the crystals, the zeolites show no evidence of isomorphous replacement under valence-control (so that 2Na replaces Ca, or *vice versa*), but, contrary to the theories of all authorities, they are either of fixed composition, like natrolite and mesolite, or they vary in composition exactly as the feldspars do; that is, variation occurs by replacement of Ca by Na (or K) with simultaneous replacement of Al by Si. The isomorphous molecules are composed of the same number of atoms, and these are divisible into three groups of atoms, the smallest oxygen atoms, the intermediate Al and Si atoms, and the largest Ca and Na atoms; in all zeolite molecules the oxygen atoms are just twice as numerous as the (Al+Si) atoms, while the number of Ca+Na atoms is constant in each part of the zeolite group whose molecules are capable of forming mix-crystals, but differs widely in different parts of the whole group.

The zeolites differ from the feldspars (1) in forming many isomorphous systems rather than one such system, (2) in apparently showing a limited range of formation of mix-crystals rather than an unlimited range, and (3) in consisting of very complicated molecules instead of very simple ones.

17. This theory of the isomorphous relationships of the zeolites can be tested by comparative studies of the composition of zeolites under varying conditions of composition of the solutions surrounding them. If the theory is correct, it can be used to learn something about the nature of solutions in given regions.

It seems very strange to the writer that Tschermak, the able exponent of isomorphous systems in silicates and the scientist who established the correct explanation of the composition of the feldspars, was unable even after years of elaborate studies and calculations[14] to discover that a similar explanation should be applied to the zeolites, and recently presented instead a theory which is so complicated that it seems destined to fall of its own weight.[15]

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[14] See his elaborate discussion of the zeolites: *Sitz. Akad. Wiss. Wien*, **CXXVI,** (1917), pp. 541-606 and **CXXVII,** (1918), pp. 177-289.

[15] While this is the view of E. T. Wherry (*Am. Min.*, **VIII,** 1923, p. 122) and the writer, it should in fairness be added that Tschermak's theory is discussed favorably by Doelter (Hdb. Mineralch. **II,** 2, 1917 and **II,** 3, 1921) and by Niggli (*Zeit. Kryst.*, **LVII,** 1923, p. 647). It is not accepted nor mentioned by Groth (Mineral. Tabellen, **1921,** p. 120).

SULPHATE MINERALS FROM WEATHERING OF SHALE NEAR ITHACA, NEW YORK

James H. C. Martens, *Cornell University*

Epsomite, gypsum, and alunogen formed by the weathering of shale are fairly common near Ithaca, and the object of this brief note is to describe their occurrence, which is probably duplicated in many other regions of sulphide-bearing shales.

These minerals are by no means confined to one geological formation, since they are found on the Moscow shale, Genesee shale, and the lower part of the Portage; but they are perhaps most abundant on the Genesee shale between Esty's and Portland Point on the east side of Cayuga Lake. There are similar occurrences in some of the gorges near Ithaca. The efflorescence of epsomite on the cliffs along the east side of Cayuga Lake near the south end was mentioned by L. C. Beck.[1]

The epsomite occurs as a loose, white, powdery coating which forms on the cliffs irregular patches often 10 sq. m. (100 sq. ft.) or more in area during dry seasons. It is thickest where a small ledge overhangs a recess, and in these places sometimes forms fluffy masses as much as 2 or 3 cm. (1 in.) thick. Microscopic examination shows the epsomite to consist of very fine curved fibers which themselves are aggregates of extremely small crystals. In the winter, due to slow recrystallization of the material, the epsomite was found in somewhat more compact form and consisted of distinct crystals up to about 0.5 mm. long by 0.1 mm. wide. The mineral was identified by its optical properties, taste, and qualitative chemical reactions.

Gypsum occurs both as a coating on the edges of the beds and between the laminae of the shale. Being less easily soluble than the epsomite and alunogen, more of it remains during the wet seasons of the year. The surfaces of the thin plates into which the weathered shale easily separates are often half covered by gypsum, in distinct crystals up to 1 or 2 mm. long, strongly colored by limonite. Small amounts of gypsum also occur mixed with the epsomite. Secondary gypsum, such as is described here, is easily distinguished from primary gypsum deposited with the shale since the latter would be more intimately mixed with the clayey material and would occur in the fresh as well as in the weathered rock.

Special conditions seem to have been necessary for the formation of the alunogen since it is much less common than the epsomite and gypsum. Perhaps, in places where it was not formed, all of the sulphuric acid united with substances more easily attacked than the hydrous aluminum silicates from which the aluminum must have come, or the acid was not of sufficient concentration to attack these aluminum compounds. The alunogen occurs as white to slightly yellow botryoidal masses of finely fibrous structure. Single crystals are rarely more than 0.005 mm. in width by 0.2 mm. in length so that aggregates are usually seen in examining the crushed mineral under the microscope. The alunogen sometimes cements fragments of shale together. The mineral was identified by qualitative chemical reactions, refractive index of 1.477 (±.005), inclined extinction and low double refraction.

Melanterite, in somewhat altered form, with refractive indices higher than those of the pure unaltered mineral, was found in small amounts at only one locality, where it was associated with the three other sulphate minerals mentioned.

The sulphate in all of these minerals comes from the oxidation of the pyrite and marcasite in the shales; the lime and magnesia of the gypsum and epsomite, respectively, are presumably from carbonates, and the alumina of the alunogen from the clayey material. The salts are brought to the surface in solution in water which evaporates and leaves the minerals as described. Secondary minerals containing the alkali metals were not found in this region.

It may very well be supposed that the formation and crystallization of these sulphates, in itself a stage in the chemical weathering of the shale, may help also in the mechanical disintegration of the rock by opening wider the fine cracks already existing, in much the same manner as freezing water.

[1] Mineralogy of New York, p. 251 (**1842**).

PROCEEDINGS OF SOCIETIES

NEW YORK MINERALOGICAL CLUB

Regular Monthly Meeting of January 21, 1925

A regular monthly meeting of the New York Mineralogical Club was held in the East Assembly Room of the American Museum of Natural History on the evening of January 21, at 8:15 p.m. The Vice-president, Mr. George E. Ashby, presided, and there was an attendance of 28 members.

The following names were presented for membership by the recording secretary and were transmitted to the membership committee for report at the February meeting: Miss Grace M. Carhart, Hunter College, New York City; Mr. C. C. Lawson, Yale University, 124 Prospect Street, New Haven, Conn.; Mr. Frederick A. Sach, c/o Hallgarten & Co., 44 Pine Street, New York City.

The chairman spoke of the death, on January 2, of Dr. Daniel S. Martin, one of the original members of the Club, and one through whose activities the interest in local mineralogy has profited through the many years of his life. It was moved by Mr. Stanton that the Chair appoint a committee to draw up a suitable resolution upon the death of Dr. Martin. In seconding this motion the recording secretary asked that Mr. Ashby be included on this committee. The motion being carried the Chair appointed Dr. Kunz and Messrs. Ashby and Stanton to serve as a committee on the Martin Resolution.

Mr. Wintringham drew the attention of the Club to several recent papers of importance including one by Dr. Henry S. Washington on the Composition of the Earth's Crust, by Hodges on a Proposed Classification of the Igneous Rocks, and by Dr. E. V. Shannon on the Mineralogy and Petrology of the Triassic Diabase of Goose Creek, Virginia. Dr. F. I. Allen drew attention to the recent death of Dr. Horace L. Wells of Yale University, and spoke of some of his achievements in science as well as of his personality. The chairman then introduced the speaker of the evening, Dr. Paul F. Kerr, of Columbia University, who read a paper on "*X-rays in the Determination of Ore Minerals.*" Dr. Kerr outlined the problem of the identification of metallic ore minerals in polished slabs and pointed out how the use of the X-ray has become a factor of importance in the determination of small amounts of metallic minerals. As a source of X-rays he described the Coolidge tube with a molybdenum anode. He explained the powder method and described the apparatus which he has adapted to the research work on the metallic minerals at Columbia. Using this apparatus the powder method can be applied to small amounts and a number of samples can be tested at once. This is an important factor since the time of exposure required varies from 12 to 20 hours.

In the discussion it was asked if the method was practical for mixtures. The speaker replied in the affirmative. Mr. Radu asked for a statement of the difference in method between the powder method and that devised by the Braggs. The speaker explained at length using concrete examples.

A vote of thanks was tendered to Dr. Kerr for his highly original and interesting paper. The recording secretary drew attention to the first minute book of the Club which, under the date of May 21, 1889, recorded the first meeting, among the names of those present being that of Dr. Martin. The meeting adjourned at 9:40 p.m.

HERBERT P. WHITLOCK, *Recording Secretary.*

NEWARK MINERALOGICAL SOCIETY

The seventy-third regular meeting of the Society was held in April with President Miller presiding. Eighteen members were present. A letter from Ward's inviting the Society to visit Rochester on May 30 was read and the secretary instructed to reply that the Society regretted its inability to avail itself of the invitation.

The secretary was instructed to obtain estimates on having the By-Laws printed and also a membership list. Mr. Broadwell reported that his list of Franklin, N. J., minerals would be ready for distribution at ten cents per copy at the May meeting.

The application of Mr. E. A. Maynard, of Jamaica, L. I., was reported favorably by the membership committee and election followed. Ten members had on display the three minerals that were the most interesting to them and were given five minutes to explain why.

Mr. Walther spoke on native iron crystals from California, pseudomorphous quartz and dendritic sphalerite; Mr. Bates, on sentimental and intrinsic values of three quartz specimens; Mr. Thowless, on Ellenville quartz; Mr. Carpenter, on quartz and prehnite from Paterson. Mr. Rankin referred to his three minerals as the three milestones in his collecting experiences. Mr. Grenzig spoke on calcite basal planes and his dream leading to his finding an amethyst specimen. Mr. Broadwell referred to schallerite from Franklin, N. J., and sarkinite from Sweden and their similarity; Mr. Karlson, on a new and unnamed mineral from Sweden; Mrs. Miller, on aurichalcite, cobalt bloom and amazonite from Valhalla, N. Y.; Capt. Miller, on the physical properties of smithsonite-willemite, uraninite and quartz inclusions in mica. WM. H. BROADWELL, *Secretary.*

PHILADELPHIA MINERALOGICAL SOCIETY

Academy of Natural Sciences of Philadelphia, April 9, 1925

A stated meeting of the Philadelphia Mineralogical Society was held on the above date, presided over by the vice-president, Mr. Trudell. Twenty-eight members and eleven visitors were present.

Dr. F. A. Cajori, of Bryn Mawr, Pa., was elected to membership. Mr. Cienkowski and others exhibited quartz crystals and limonite geodes from Henderson, Pa., and its vicinity. Messrs. Boyl, Arndt, and Clay exhibited natrolite from Moore, N. J.

The program of the evening consisted of a series of short talks on various topics of mineralogical interest by different members of the society. Mr. M. J. Biernbaum first discussed "*Mineral Freaks.*" Some striking characteristics of certain minerals were illustrated by appropriate specimens. The improvement of crystal specimens by the careful removal of the surrounding matrix was described by Mr. H. W. Warford. Specimens were exhibited illustrating the results of both legitimate and illegitimate "development" of this kind. Mr. G. Faust and Mr. F. Speer told of the growth of the interest in mineralogy aroused by members of the society among the students of the Northeast High School of Philadelphia, and described the present activities of the students in this science.

Mr. H. E. Millson then demonstrated a number of simple chemical tests by which various minerals can be distinguished in the field. With but few reagents and little apparatus a number of minerals similar in appearance may be readily identified. Mr. H. R. Blank concluded the program with a discussion of the use of the flame colorations given by various elements as an aid to the identification of minerals containing them.

Horace R. Blank, *Secretary.*

PHILADELPHIA MINERALOGICAL SOCIETY

Academy of Natural Sciences of Philadelphia, May 14, 1925

A stated meeting of the Philadelphia Mineralogical Society was held on the above date, with the president, Mr. Vaux, in the chair. Thirty-one members and seven visitors were present.

Mr. Oscar Schuck of Philadelphia was elected a junior member, and Messrs. George Urban and Charles R. Lorimer were proposed for junior membership. A trip to view the collection of Mr. Canfield, of Dover, N. J., was announced for May 24th.

Mr. Keeley announced the death, on April 19, 1925, of Mr. George W. Fiss, of Philadelphia. Mr. Fiss was a pioneer in the collection of mineral specimens for study under the microscope, and his enthusiasm in this direction was of great influence in promoting the use of this instrument among mineral collectors.

Mr. M. J. Biernbaum addressed the society on "*Crystal Twinning.*" The different types were defined, and the distinctions between contact and penetration twins were illustrated. Some of the common laws of twinning in the different crystal systems were illustrated by means of drawings and models.

Mr. J. C. Boyle then discussed "*Methods of Mineral Study,*" and pointed out the advantages of correlating minerals according to their occurrence and manner of formation. A knowledge of mineralogy from this standpoint is of great value to the collector in the field.

Mr. George Vaux, Jr., described his recent visit to the gem mineral localities of southern California. Very fine pink and green tourmaline and colorless beryl crystals from the Pala district were exhibited; also very beautiful transparent lilac crystals of kunzite over 10 cms. long. California garnet, lepidolite, topaz, benitoite, and neptunite were also shown.

Horace R. Blank, *Secretary.*

DOUBTFUL SPECIES.

CLASS: SULFO-SALTS. DIVISION: $RS:Bi_2S_3 = 5:3$.

Wittite

K. Johansson: Ett par selenförande mineral från Falu Gruva. (Two selenium bearing minerals from the Falun Mine.) *Arkiv Kemi, Mineral., Geol.,* **9,** No. 9, p. 2 (1924).

Name: In honor of the Swedish mining engineer, Th. *Witt.*

Chemical Properties: A sulfo-bismuthide of lead. Formula, $5PbS. 3Bi_2(S, Se)_3$. Analysis: Ag 0.19, Pb 33.85, Cu 0.08, Fe 0.28, Zn 0.26, Bi 43.33, S 12.14, Se 8.46, insol. 0.54; sum 99.13.

Crystallographic Properties: X-ray diagrams of this mineral indicate an orthorhombic or monoclinic symmetry.

Physical Properties: Color light lead-gray. Streak black. Cleavage good. Luster metallic. H=2-2.5. Sp. gr. 7.12.

Occurrence: Found at the Falun Mine with dark gray quartz and magnetite in a cordierite-bearing amphibole rock.

Discussion: Apparently a new mineral but lack of metallographic examination of the analized material makes it uncertain if the selenium belongs to this mineral or is intermixed as has been found to be case with other Falun minerals.

W. F. Foshag.

CLASS: SULFIDES, ETC. DIVISION: R : S=1 : 2.

Cobaltpyrite

K. Johansson: *Op. cit.*; this mineral p. 2.

Name: In allusion to its composition, a *cobalt* bearing *pyrite.*

Chemical Properties: A sulfide of iron and cobalt. Formula, $(Fe, Co)S_2$. Analysis: Fe 33.32, Co 13.90, Ni 0.19, S 52.45; sum 99.86. Ratio of Fe:Co=0.597: 0.236.

Crystallographic Properties: Isometric, octahedrons.

Physical Properties: Color like pyrite but tending toward brown. Luster metallic. Cleavage octahedral. Fracture conchoidal to splintery. H. 6. Sp. gr. 4.965.

Occurrence: Found in cavities with quartz in crystals up to 3 cm. across.

Discussion: The ratio of iron to cobalt is high and the mineral should therefore be considered a cobaltiferous pyrite. This term would be more appropriate than cobaltpyrite. W. F. F.

"Borgstroemite"

Martti Saxén: The Petrology of the Otravaara Region of Eastern Finland. *Bull. Com. Geol. Finland*, No. 65, p. 50-55, **1923.**

Name. In honor of the mineralogist, L. H. *Borgstroem.*

Chemical Composition: A basic sulfate of iron, $3Fe_2O_3 . 4SO_3 . 9H_2O$. Analysis: Fe_2O_3 50.55-51.83; SO_3 28.73-29.41; H_2O 19.44-20.04. Insoluble in water, soluble in acids.

Physical Properties: Color yellow. Earthy.

Occurrence: Forms a zone between the limonite capping and the pyrite ore bodies at Otravaara, Finland.

Discussion: The general description and occurrence suggests a member of the jarosite group, especially the member carphosiderite. The formula assigned is that of a compound found by Posnjak and Merwin (*Jour. Am. Chem. Soc.*, **44**, 1977, 1922). The agreement with the theoretical composition, however, is not good. The mineral should tentatively be referred to carphosiderite.

W. F. F.